Foreign and domestic contributions to surface ozone in Spain

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EXTENDED ABSTRACT

Tropospheric ozone (O_3) exerts strong adverse impacts on human health, climate, vegetation, biodiversity, agricultural crop yields and thus food security. O_3 is formed in the atmosphere through non-linear photochemical reactions involving volatile organic compounds (VOCs) and nitrogen oxides (NOx) precursors [1]. Furthermore, meteorological stagnation, high solar radiation, high temperatures and low precipitation favor the formation of tropospheric O_3 at surface levels exceeding target regulatory values [2]. Due to the complex and poorly constrained physico-chemical O_3 formation and removal pathways, no straightforward strategies currently exist for reducing O_3. Currently, there are no observational methods that differentiate the origin of O_3. Despite their inherent uncertainties, chemical transport models (CTMs) allow for the apportionment of the contribution of any source to O_3 concentrations. The mass-transfer source apportionment method is an optimal approach to study the contribution of different sources to ozone levels [2].

In this study, we provide a quantitative estimation of the foreign and domestic contributions to surface ozone on Spain, relative to European countries and the contribution of hemispheric background ozone. For that, we use the CMAQ-ISAM within the CALIOPE air quality modelling system to simulate the O_3 dynamics over Europe quantify national contributions for the ozone season from May to October in 2015. We tag both O_3 and its precursors, NOx and VOCs, from the different European countries, all the way through their lifetime, from emission to deposition.

A. Air quality model and simulation set-up

The CALIOPE air quality model [3] was used to simulate the surface ozone concentration over Europe. CALIOPE integrates a meteorological model (WRF-ARWv3.6), an emission model of anthropogenic emissions (HERMESv3) [4] and biogenic emissions (MEGANv2.04) [5], and a chemical transport model (CMAQv5.02) [6]. For the present study, the system is configured with a domain over Europe at a resolution of 18 km x 18 km and 37 vertical levels (top of the atmosphere at 50hPa) and uses meteorological and chemical boundary conditions from the ERA-interim and CAMS analysis, respectively. CMAQ uses the gas phase mechanism CB05 with active chlorine chemistry and toluene mechanism (CB05TUCL) [7] and the sixth generation CMAQ aerosol scheme.

The Integrated Source Apportionment Method (ISAM) within CMAQ allows us to quantify the contribution of O_3 precursors from different countries to the O_3 surface concentration over the European domain [6]. The CMAQ-ISAM tracks both the O_3 and its gas precursor emissions, NOx and VOCs, from the different European countries, boundary conditions and initial conditions. Each tagged species is subject to the physical and chemical processes that it might undergo in the atmosphere. In this study, we tracked the contributions of 35 European countries, the non-European countries, the sea, the chemical boundary conditions (BCON) and the initial conditions.

We focus the study over the period May to October 2015. The summer of 2015 was a hot summer in Europe compared with the reference period 1981-2010; it was especially hot in central Europe and in the Mediterranean. In Spain, there was a heatwave of 26 days during the month of July, making the year 2015 of particular interest to understand O_3 formation.

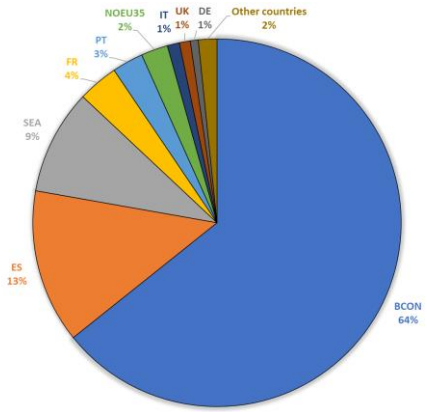

Fig. 1 Foreign and domestic contributions to the MDA8 O3 in 2015 (May-October) in Spain.

B. Results

The maximum daily 8 hours averaged (MDA8) O_3 concentration in Spain for the period of study is 116.10 µg/m^3. In figure 1, we show the relative contribution of the different tagged sources to the MDA8. The imported O_3 is by far the most significant contributor to the ground level O_3 concentration (86 %) and only 14 % is coming from the emitted precursors in Spain. From the foreign contributions, the BCON plays a key role in the O_3 concentrations in Spain. This result shows the importance of the hemispheric and intercontinental transport of O_3 background in the concentrations of Spain. The emissions coming from the sea contribute an overall 10,71 µg/m^3, being the third most important contributor in Spain. Although ship emissions mainly affect coastal regions, their impact on O_3 concentrations is close to the sum of the main country emissions

The contributions of individual European countries to O_3 concentrations in Spain remain below 4%. France and Portugal are the main countries affecting Spain (4 % and 3 %, respectively), and the sum of the other countries contributes a total of 7 %.

Figure 2 shows the MDA8 O_3 surface concentration over Europe for our period of study due to the tagged species (emissions) from Spain. Higher concentrations are found over Spain, but Spain contributes to the O_3 surface concentrations of other European countries, as Portugal and France, clearly affected by proximity. Spain contributes a total of 8.5% and 3.5% of the total ozone concentration in Portugal and France, respectively. Interestingly, the balance between neighbor countries is quite different. Spain is exporting O_3 potential formation to Portugal while importing it from France. In relation to the other European countries, the contribution of Spain is nearly negligible.

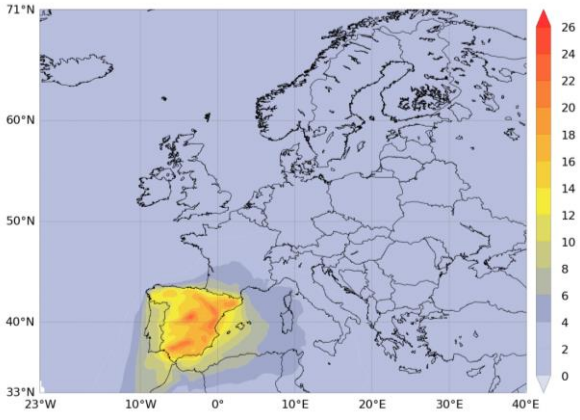

Fig. 2 MDA8 O3 surface concentration over Europe in ug/m3 due to the Spain emissions.

The most polluted regions in Spain are the air basins of Madrid and Barcelona, due to the respective emissions of O_3 precursors in the main urban areas. But the south of Spain also shows important O_3 levels, up to 130 µg/m^3 on MDA8 O_3 average in the studied period. Concentrations there are higher than in Madrid and Barcelona. Note that the south of Spain is not strongly affected by the O_3 contributions from other countries, despite the non-negligible contribution of the North African countries and ships emissions. Our results indicate the importance of understanding the source of the pollutants and how the topography and meteorological conditions play an important role in the O_3 surface concentration. This information is a fundamental step to design effective policies to reduce O_3 in a country and quantify the margin of action that local actions might have.

C. Conclusions and future work

Our study has provided a first estimation of the principal contributors to the O_3 surface concentration in Spain during the ozone season in 2015. The results show the importance of the imported O_3 over Spain and point out the relevance of the long-range transport. To achieve the European air quality standards in Spain and avoid the exceedances of the O_3 target values, it is necessary to decrease not only the domestic emissions of O_3 precursors, but also the emissions from the intercontinental transport, the shipping transport, and the other European countries. So, the regulatory efforts have to be designed as a combination of local and foreign emission reduction actions.

Future work will extend this analysis to other European countries and different years to provide a general picture of the balance between the foreign and domestic contributions to surface ozone among Europe. Furthermore, we plan to combine these results with health information and advance our understanding of the impact of O_3 upon the mortality burden in the continent.

D. ACKNOWLEDGEMENTS

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Author biography

Roger Garatachea Solé received his BSc degree in Chemistry from the University of Barcelona (UB) in 2018. In 2021, he completed his MSc degree in Environmental Engineering at Polytechnic University of Barcelona (UPC), and he did his master's thesis at the Barcelona Supercomputing Center (BSC). In September 2021, he enrolled the Atmospheric Composition Group of BSC as a Junior Researcher Engineer.